



BEST AVAILABLE COPY

PATENT

I hereby certify that this paper or fee is being transmitted sufficient postage as Express Mail No. EV 531802399 US, addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on January 14, 2005.

Dated: January 14, 2005

BY: _____
Rodney D. DeKruif

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: David C. Dunand)
)
Serial No: 10/680,639)
) Attorney Docket No. 6513-DIV
)
Filed: October 7, 2003)
)
For: SUPERCONDUCTING)
Mg-MgB₂ AND RELATED)
METAL COMPOSITES)
AND METHODS OF)
PREPARATION)

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

RULE 131 DECLARATION OF DAVID C. DUNAND

1. I, David C. Dunand, am inventor of the invention (the "Invention") disclosed and claimed in the above-entitled application (the "Application"). I am a Professor in the Department of Materials Science and Engineering at Northwestern University. I make this declaration in support of the Application and, in particular, to antedate a reference cited against the Application.

2. The Invention claimed in the Application was completed before the effective date of the journal article by Sharoni, et al., entitled "Spatial variations of the superconductor gap structure in MgB₂/Al composite," *J. Phys. Condens. Matter* 13 (2001) L503-L508 (*i.e.*, the Sharoni reference). More specifically, the

Invention was conceived and with due diligence reduced to practice prior to the effective date of the Sharoni reference. (The effective date of publication, i.e., June 4, 2001, can be found at <http://www.iop.org/EJ/toc/0953-8984/13/22>.)

3. This Declaration, and prior invention, is supported by copies of pertinent pages from my laboratory research notebook, entries which were contemporaneously witnessed by Graduate Student Naomi Davis. Date redacted copies of the aforementioned notebook pages are provided collectively as Exhibit A and incorporated herein by reference. These documents establish that the Invention was made at least as early as March 26, 2001, which is a date earlier than the effective date of the Sharoni reference.

I hereby declare that: All statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; that those statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code; and that willful false statements may jeopardize the validity of the Application or any patent issuing thereon.

Date January 13 2004

David C. Dunand
David C. Dunand

Monday

I discussed last week with D.N. Seidman the idea of superconducting composites based on the new compound MgB_2 . I had mentioned to him the following simple idea:

(1) Infiltrated Mg - MgB_2 composites

The Mg - B phase diagram shows that Hg and MgB_2 are at equilibrium with each other. It should thus be possible to infiltrate a packed bed of MgB_2 powder (which may have been pre-sintered to make a continuous skeleton) with liquid Hg . The net result is a composite with a continuous superconducting phase of MgB_2 , embedded in a continuous metallic Hg phase, which allows for thermal management (conducting heat away from MgB_2 and preventing loss of superconductivity) and also being able to carry current if s.c. is lost.

The following other ideas came to me, undisclosed to D.N. Seidman:

(2) Fabrication of MgB_2 fibers

Superconducting fibers would be very useful embedded in a non s.c. matrix (polymer, metal, glass, ceramic) which provides thermal and mechanical properties. Existing s.c. fibers can be made by drawing (when ductile) or by powder sintering (when brittle). My idea uses a different technique, i.e. the Taylor wire technique.

MgB_2 powder are packed in a "glass" tube ("glass" any amorphous ceramic such as Pyrex, E-glass, etc). The MgB_2 is melted and the glass is quickly drawn into a hollow fiber containing MgB_2 which solidifies into a continuous fiber, which can be spooled

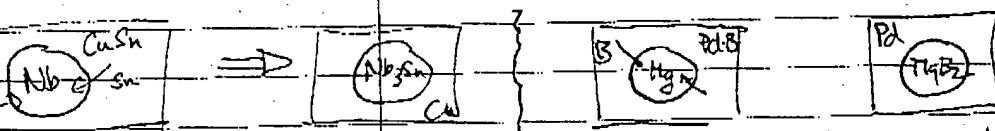
as shown. (Schematic diagram of the process is understood N.G.)

Taylor wires have been made with many metals (see Donald, 1987).
 The following requirements are needed: (1) no reaction between glass and MgB_2 , (2) working temperature of fiber must be higher than MgB_2 (3) glass must become highly viscous before MgB_2 diffuses, lest it deforms and breaks the MgB_2 . Pyrex seems a good first choice.

The glass sheathed MgB_2 fiber can then be embedded in a matrix (metal, glass, ceramic, polymer) to form a composite by usual composite processing techniques. Alternatively, the glass can be dissolved by an acid (HF-containing acid) and the bare MgB_2 fibers can be used in a composite. Some dissolution of MgB_2 may be tolerable as part of the glass removal.

(3) Ductile $Mg-B-Pd$ wires as precursors for $Pd-MgB_2$ composites

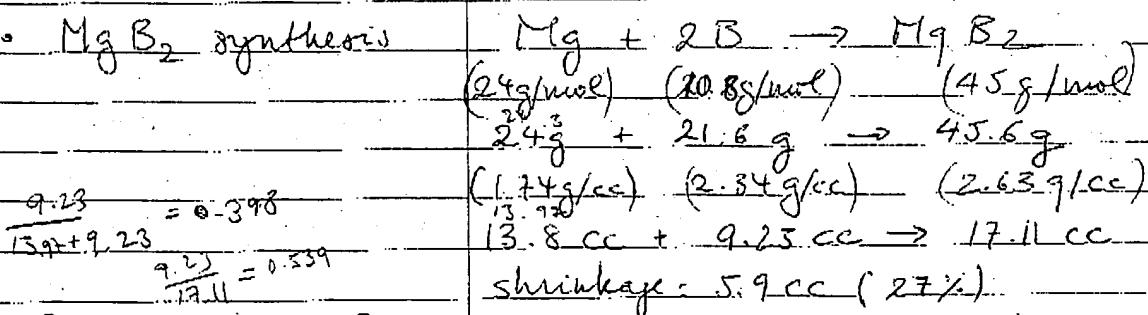
Nb_3Sn-Cu composites are ideal, as Cu prevents thermal run-away. They are produced by stacking Nb wires between a Cu-Sn bronze matrix and drawing, resulting in a ductile Cu-Sn/Nb composite which can be drawn and shaped (coiled, etc.) to shape. Heat-treatment diffuses Sn to the Nb fibers, forming the brittle Nb_3Sn superconductor in a Cu matrix.



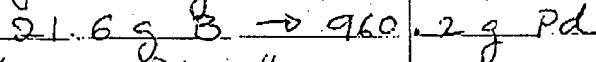
The same idea can be used for MgB_2-X composites, where X is a ductile metal which can be alloyed with B. A good example is Pd, which can accept 2.2 wt% B (at%). So, Mg wires could be embedded within sheets of Pd-B alloy, diffusion-bonded, drawn or shaped as needed. Upon heating below 650°C (melting point of Mg),

calculations for volume fractions of Pd/MgB₂ composite

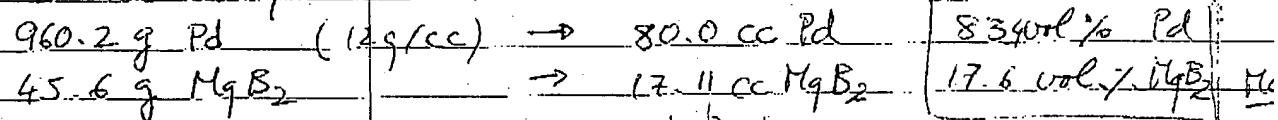
• MgB₂ synthesis



• Pd-B alloy Pd-2.2wt% B (max solubility)



• Mg-B-Pd alloy



→ 1/6 of the volume is MgB₂

$$\frac{\pi a^2}{4b^2} = 0.176 \quad \frac{a}{b} = \sqrt{0.176 \frac{4}{\pi}}$$

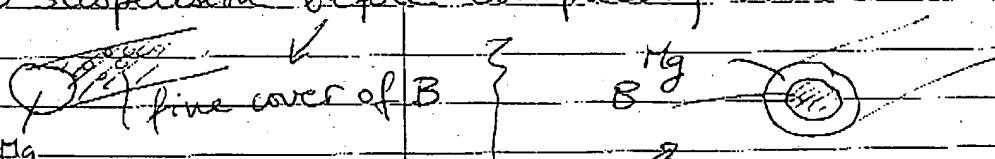


a

This volume fraction is not very high, but still commercially feasible, provided price of Pd is not too high.

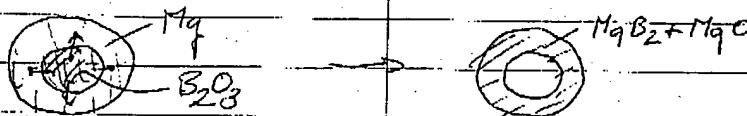
N.B.: some Mg will dissolve in Pd

- Additional B could be introduced by dipping Mg wires in B suspension before compacting with Pd or Pd-B foils

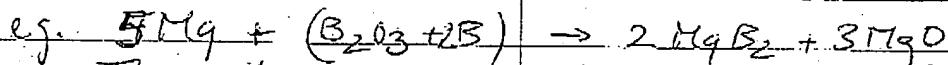


or filling Mg tubes with B powder and then compacting with Pd. Extrusion would still be easy with B powder
and understood Hartwig, Liao

- Alternative is to fill Mg tubes with B_2O_3 , which is viscous at extrusion temperatures. The reaction is then



or a suspension of B particles in B_2O_3 , which will remain viscous & drawable.



The advantage is that the metallic matrix can now be a metal different from Pd, and the volume fraction of fibers has no restrictions

MgP₂ fibers (160 μm in Ø) have been disclosed at the APS Fall meeting: B fiber is exposed to Mg vapors and form MgB₂ which was found to be superconducting.

Taylor wire technique has following advantages:

- much more rapid production
- diameter easily changed (1-100 μm typically)
- different grain structure
- sheath of glass

Good matrices for "filled-tube" process: any metal which does not dissolve into Mg (which would contaminate MgB₂)

1. Cu : zero sol. in Mg ; eutectic at 48°C; $MgCu_{2.3}Mg_2Cu$; 22wt.% Mg in Cu
2. Ni : zero

read → understood,

1 form of Paris

Solubilities

Metal X	sol. in Hg	Begins at	Intermetallics	Fus. temp. (°C)
Cu	0	~3 wt%	2	483
Ni	0	0	2	506
Pt, Ir	0.12	~1.5 at%	0	651
Ta, Co	0	0	1	635
Fe	0	0	0	—
Cr	0	0	0	—
Al	0	huge	many	575
Be	0	0	1	—
Pd	0.23	5 wt%	7	540
Pt	0	0	5	575
Nb	0	0	0	—
Mo	0	0	0	—

Other advantages:

high conductivity : (Cu) (but reduced by Hg in solid soln)

Au (but %)

Processing : (Cu) low melting

electrodeposition : Cu, Ni, Cr, Au, Pd, Pt

no borders (Cu), Au

read & understand

Naomii & Lai

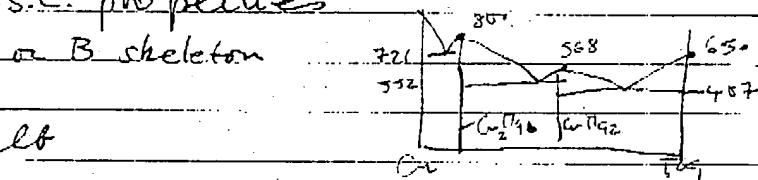
Other idea:

React B with Cu-Mg melt $2B + Cu + Mg \rightarrow MgB_2 + Cu$
this is only possible if Cu does not dissolve and
deteriorates MgB_2 s.c. properties

1. infiltrate B fibers
with Cu-Mg melt

2. react in the melt

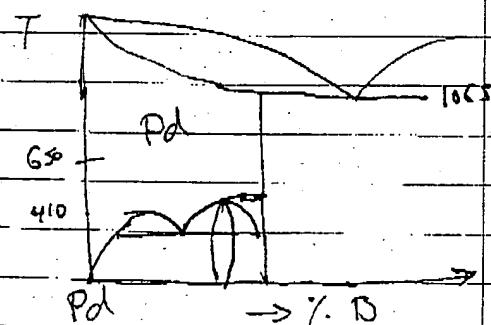
3. solidify Cu-MgB₂ composite.



This will work with non-bride formers, i.e. mostly Cu and the

Read, understand

of binary phase diagram



pure B dissolves Cu ($\sim 16\text{ wt\%}$) at 1000°C

$$1\text{ mol B} = 4.61\text{ cm}^3 \sim 40\text{ vol\%}$$

$$\frac{1}{2}\text{ mol Mg} = 6.89\text{ cm}^3 \sim 60\text{ vol\%}$$

$$\begin{cases} 30\text{ vol\% B} \\ 45\text{ vol\% Mg} \\ 25\text{ vol\% Cu} \end{cases} \rightarrow \begin{cases} \sim 50\text{ vol\% MgB}_2 \\ 2/3 \text{ MgB}_2 \\ 25\text{ vol\% Cu} \end{cases} \rightarrow \begin{cases} 1/3 \text{ Cu} \end{cases}$$

BMG → batch program works! → code

some cryst. matrix! → new paper with double
no creep error due to self-loading after
2 h hold → throw away data

stand alone { - compression
paper { - hold
matrix info

• MRS paper → draft on Monday
practice talk on Thursday

task (avr Dearn) (cont)

↳

↳ ~~at~~ carbon steel

Taylor wire MgB_2 trial

(Went to glass blowing shop and tried with pyrex (10mm))

- can draw easily empty wire

- MgB_2 powder (~ 1cm high) gets red hot, but no melting visible
Can partially draw pyrex, but eventually fracture. MgB_2
does not burn.

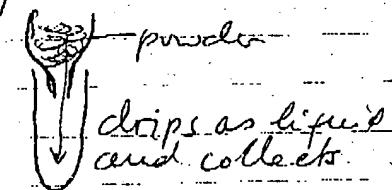
Problem is φ is too large, cannot keep the whole block
of MgB_2 hot. → try finer tube, maybe try a torch
with multiple outlets

Tried finer wires ~ 4 nm, and after a few trials, got a wire ~ 100-200 nm, with black MgB₂ inside. It is not clear whether the powder was melted throughout, but it seemed to be

- tried Cu in 4 mm tube (pyrex), but not successful due to high T_m (glass cracked before Cu melted)

- Zn powders worked, nice Taylor wires.

- Tried again MgB₂ and got a nice length ~ 10 cm uninterrupted. Pyrex was not cleaved, so bubbles interrupt wire in some parts. Maybe also need to "pre-melt" materials, e.g. ~~Mg~~ powder as done for AgCl during thesis (but T_m = 455 °C vs 800 °C for MgB₂)



glass shop closed

read Campbell paper (PRL 86, 2423) and had the following idea: integrated fiber formation & composite fabrication

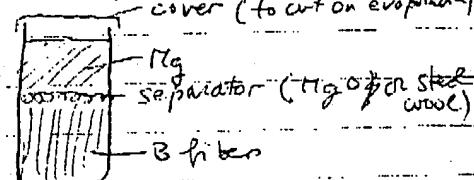
cover (to cut on evaporation)

1. Put B fibers and Mg into crucible

2. Heat at 950 °C → Mg melts and seals crucible

hold for 2 h → Mg vapors react with B fibers and form MgB₂ fibers like

Campbell's paper



3. Pressure with gas and force Mg into fiber preform

→ MgB₂-Mg Composite

One of the main advantages is that MgB₂ fibers are formed in-situ

They do not have to be handled, bundled, etc., which would

cause damage or break them, because Campbell describes them

"... in this manner handling one heating only"

Another possibility is to infiltrate B-fibers with molten Mg and keep the composite at 950°C for 1-2 h, long enough for complete reaction to form MgB_2 . Solidifying gives a Mg composite with MgB_2 fibers.

Method to produce MgB_2 metal matrix composites

Caufield's paper shows that MgB_2 fibers can be produced by exposing B fibers to Mg vapors at 950°C for 2 h. However, the fibers are very brittle and bent after fabrication. It will be difficult to bundle them and subsequently infiltrate them without breakage. A solution is to synthesize and infiltrate the fibers in two closely consecutive steps, without handling the fibers.

Step 1: Heat-up assembly to synthesis temperature. Mg melts and vaporizes, reacting with B-fibers to form MgB_2 . If the metal I is melted, it forms a liquid seal, thus preventing escape of Mg vapors. A non-wetting separator prevents metal I from contacting fibers. Examples for metal I: Mg, Al, Cu alloy with low ~~low~~ solubility (bronze, brass).

Step 2: pressure with gas the crucible, forcing liquid metal I through the separator and between the MgB_2 fibers, thus making a composite. Solidify and extract composite from crucible. If metal I has higher melting point than synthesis temperature of fibers, first raise temperature to melt it.

Complex shapes could be made by this process. A variation is to put Mg below fibers with separator permeable to Mg vapors.

see p136 : 24) B Mg 13.8 cc Mg 60 vol\%
 $+ 21.6 \text{ g B}$ 9.23 cc B $\sim 40 \text{ vol\%}$

so at best, 20 vol\% B

$30 \text{ vol\% Mg powder}$

$50 \text{ vol\% porosity}$



$$\phi 5 \text{ mm} \quad A = 19.6 \text{ mm}^2 \rightarrow 3.7 \text{ mm}^2 \text{ B}$$

each fiber is $100 \mu\text{m} \phi \rightarrow \pi \cdot 0.05^2 = 0.2 \left(\frac{\pi}{0.1}\right)^2$
 $\{500 \text{ fibers}\}$

→ drop into $0.3"$ crucible

@ 3 cm $\rightarrow 10 \text{ m}$

Observe Dorian run infiltration.

1. Turn H₂O on (circulate first at 125 psi)
open to machine → flow switch on
zero pressure transducer
2. Close vessel - slide in
- put PB sheet
- tighten nut until lid is flush with vessel
3. Connect 1 p-transducer (showing as psix5, full vac = -65 to -70)
4. TC (check them)
5. Open gas tank, set 2nd stage regulator at 150 psi
6. purge 3 times - use valve 1 only →
check for vac. leak: insulate vessel
verify vac. is constant for 4-5 min
7. Switch on main power, turn to 50% on both zones
→ upper zone: 5.5 A / 100 V
→ lower zone: 4 A / 100 V
wait ~ 1½ h → 350°C
turn to 75% (top only)
→ upper zone: 8.5 A / 150 V
8. put T recorder on
wait ~ 45 min
wait ~ 40 min
→ turn to 90% (top only) (70% bottom)
→ turn down to 50% (bottom)
and fine-tune
9. Infiltrate 45 min
 1. increase to 550 psi (regulator)
 2. open valve #2 to vessel
 3. switch off power
 4. open valve #1 and pressurize to 500 psi
(35 atm)
 5. close valves 1 & 2 (will leak a 1-2 pressure)
10. Cool down → h to 200°C
turn off water, vent (use vent valve)
open

Mg 99.99% 2 pieces from Euema (2.6.33g)

Started cutting pieces

Cut tubing & cleaned tubing.

$$\left\{ \begin{array}{l} \text{tubes: } 0.555'' \text{ ID} = 14.10 \text{ mm} \\ 0.305'' \text{ ID} = 7.75 \text{ mm} \end{array} \right.$$

$$2 \text{ filling in small tube to } 2 \text{ cm: } V = 943 \text{ mm}^3 = 0.943 \text{ cm}^3 \rightarrow$$

assuming 50% packing: 0.472 cm^3

$$0.472 \text{ cm}^3 \approx 2.63 \text{ g/cc} \rightarrow 1.24 \text{ g MgB}_2$$

$$1.70 \rightarrow 0.802 \text{ g Mg} \quad \text{double}$$

$$\text{for large tube, multiply by } \frac{(0.555)^2}{0.305} = 3.3 \rightarrow 4.1 \text{ g MgB}_2$$

$$5.3 \text{ g Mg}$$

was (called up)

else will send ~ 30 feet of 4 mil B fibers for free

Goodfellow charges at \$100 for 10m. [minimum order: 1.16 ^{4 mil} ~~4 mil~~ \$992 for
5 mm W core, 100 mic diameter, 20.2 mg/m] \$1159 for 6 m

$$\rightarrow 10 \text{ m} = 0.202 \text{ g ...}$$

prepare small crucible: Mg: (1.605 g) (2 pieces)

add MgB₂ to crucible bottom far: 48.210 g

-629 top + push with rod

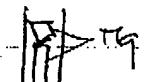
48.208

383

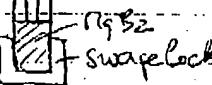
48.441 g

48.441 g

add pieces of Mg on top



↳ 1.231 g MgB₂



measured height: 6.5 mm ??
not right, was not "bottom" at first!

4.5 mm for 0.233 g → or 22.5 mm for full mass, ok!

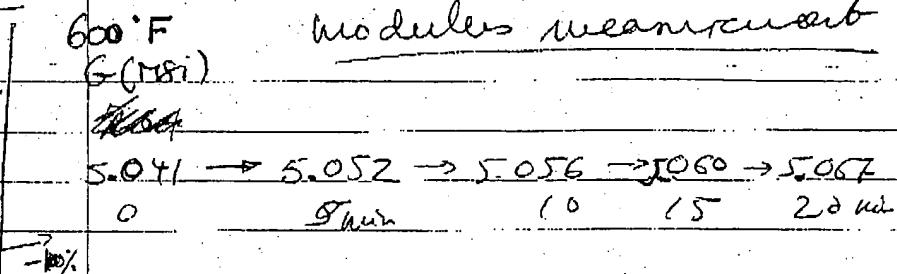
leave for cooing

146

see Abkowitz

	RT E(MPa)	500°F G(MPa)
Ti64 : 16.88	500°F	500°F
Ti64+10Ni : 15.67	5.041	5.052 → 5.056 → 5.060 → 5.067

(5.609)



-1%

- N dissolution : no, T too low, G increases
- w form of : no, " " " (check if Ar is used)
- $\beta \rightarrow \alpha$ transf. is most likely!

$$\text{mass} = 4.633 \\ \text{volume } \frac{T_{14}}{4} \cdot 1.5 = 2.37 \text{ cm}^3 \\ \Rightarrow 1.98 \text{ g/cm}^3$$

prepare large crucible 4.633 g Ti₅B₂ (15 mm height)

5.768 g Mg - 29.99% (2 large pieces, round)

- put a thin Al₂O₃ felt layer on top of Mg, stuff crucible to the top w. Al₂O₃ felt (to prevent powder blowout during arc)
- load sample in vessel, contained in S.S. crucible/pot
- evacuate/purge 3 times

11:35 vacuum -76, innulate vessel (leak check)

12:15 up low -75 → 0 K, no leaks

12:18 50% 50% T₂ T₃ T₄ vac 20.7 -76

12:30 60% 60% 191 -73

1:40 " " J16 -70

2:00 75% 75% 582 J68 -70

2:35 65% 65% 742, 733, 733, 734, 735 -69

2:43 733, 733, 733, 734, 735 -69

2:49 68% 68% 732 -69

2:55 " " 734, 732, 733, 734 -69

2 minute pressurization to 2400 (980 psi)

3:40 depressurize 170°C, 1400 (280 psi)

Mg is at level of T₂ T₄

2 crucibles
Steel crucible

147

scr

TG7
2.0 mi

20 min

(used)

$\approx 2.3 \text{ g/cm}^3$

$\approx 1.8 \text{ g/cm}^3$

lit)

(small)

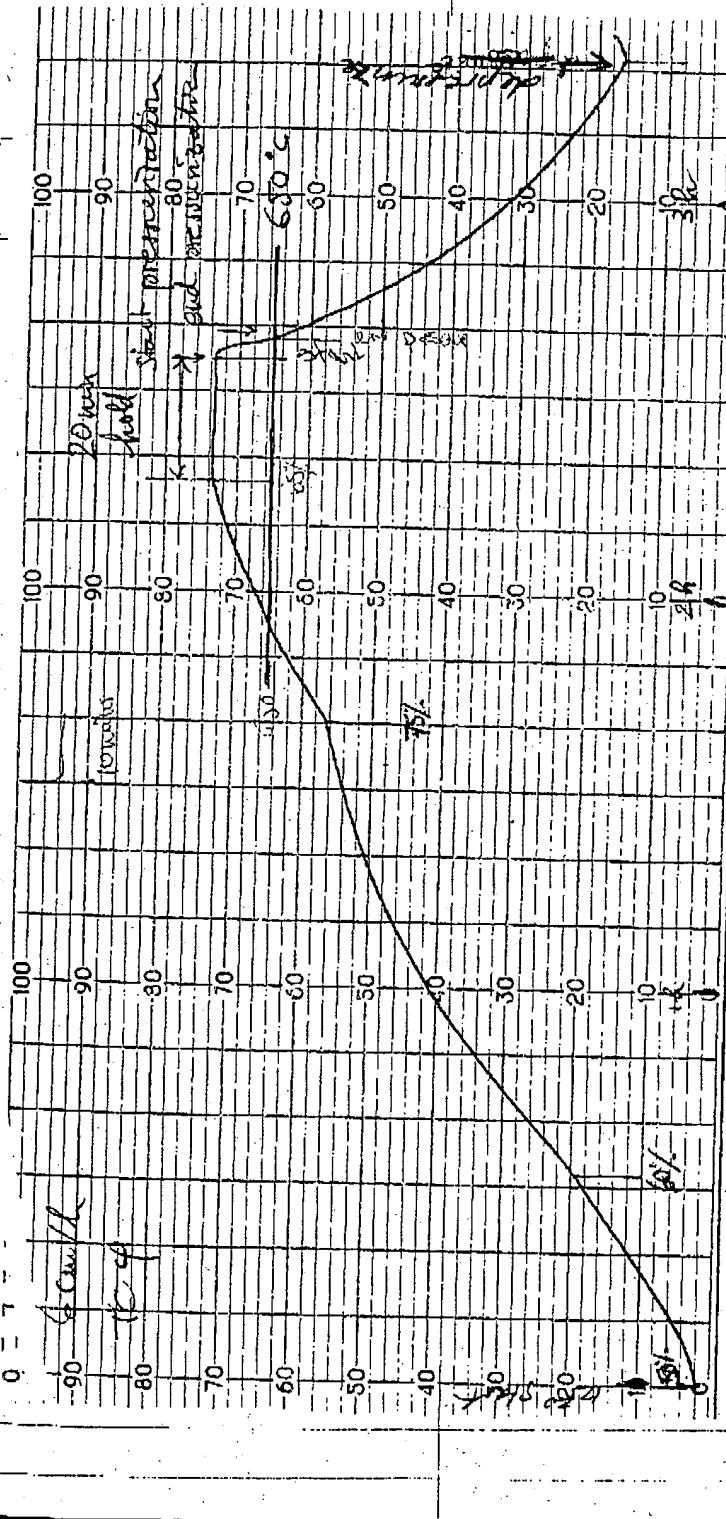
(le

wyrd)

TG4

nickles

cuvette



GRAPHIC CONTROLS CORPORATION BUFFALO, NEW YORK

PRINTED IN U.S.A.

- open at $\approx 160^\circ\text{C}$
- cuvettes are fine
- insulation is on top
- remove insulation. TG7 was melted, but level seems high \rightarrow did TG make a poor seal?
- Cut open large crucible: pieces were not melted! except for bottom piece which showed minor slumping. The time held and/or max temp. were too low.
- pour MgB_2 powder on paper \rightarrow reuse
- reuse MgB_2 pieces, except cut off top which was in contact with ZnO_2 , looks very black.
- Some white deposit in tube above Mg + possibly as reaction with ZnO_2 ?

148
939

new large crucible: old Mg B₂ powder from previous expt. 3.475 g
new 0.939

old Mg: 4.558 g 2 pieces

new Mg: 2.4704 1 piece

[4.414 g]

(Alfa Aesar)

[7.028 g] (99.99%)

add 0.3047 Mg to small crucible

both crucibles have Mg to the rim, put no felt on top

- load crucible in infiltrator

put them in flude graphite crucible
on top of s.s. crucible

- much less insulation as last time
should get better heating

TC3 is next to gr. crucible

TC4 is just above steel crucibles

- evacuate to -67, insulate & leave for the night 11:35 PM

93° AM, vac at -41, good bal! Evacuate & flush twice

P T₁ T₂ T₃ T₄ upper lower
 100 100 100 100 100 100

	P	T ₁	T ₂	T ₃	T ₄	upper	lower	
9:55	-55	24	22	19	21	70%	70%	charge Ar bottle

10:25	-55	406	382	370	295	"	80%
-------	-----	-----	-----	-----	-----	---	-----

10:47	-53	568	569	590	538	75%	85%
-------	-----	-----	-----	-----	-----	-----	-----

11:14	-51	683	702	732	712	90	95%
-------	-----	-----	-----	-----	-----	----	-----

11:30	-50	797	804	825	801	80%	95%
-------	-----	-----	-----	-----	-----	-----	-----

11:45	-49	795	795	801	799	"	80%
-------	-----	-----	-----	-----	-----	---	-----

12:05	-49			803	0	"	switch off, infiltrate
-------	-----	--	--	-----	---	---	------------------------

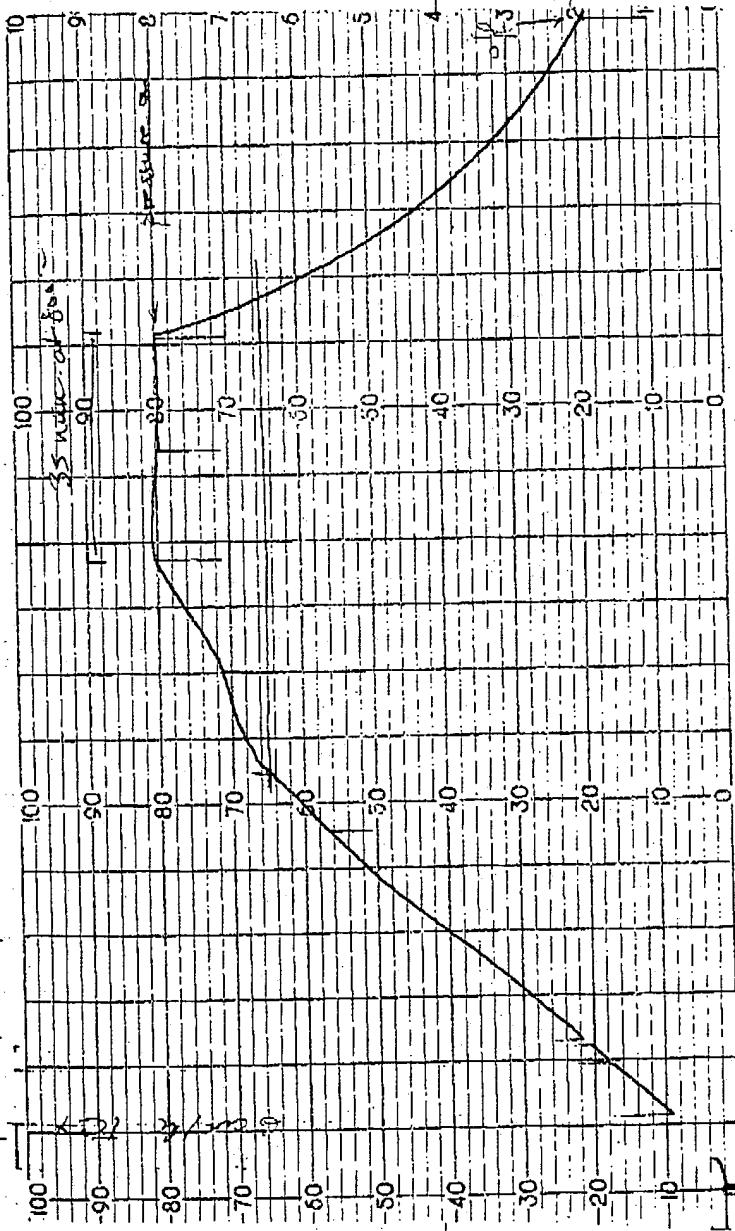
12:07	2525			755	0	0	in Goxco
-------	------	--	--	-----	---	---	----------

12:10	2232			662			
-------	------	--	--	-----	--	--	--

12:39	1468			280			
-------	------	--	--	-----	--	--	--

12:52	1312			195			pump out, open
-------	------	--	--	-----	--	--	----------------

149



PRINTED IN U.S.A.

CHART NO. 880 195-6970-00

T-305 P.017/023 F-070

JAN-12-2005 05:18PM FROM-

Carbon crucible contains 4.0 g of ~~the~~ coarse, grey powder
could be Mg evaporated and condensed?

▲ cut large crucible → Mg has melted!

small oxide "hull" left in upper part; ^{cut 1}

(very light) Some white deposit on top (MgO ?). Cut

Cut 1: hack saw, see pipe hole

Cut 2: diamond saw.

looks infiltrated!

Mg with deep central sol. pipe hull (removed)

$\xrightarrow{\text{cut 2}}$ plug

weld steel

SS

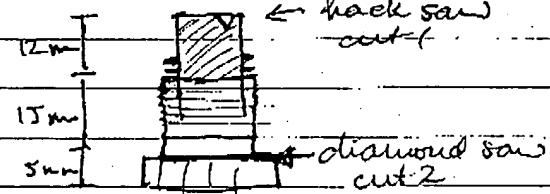
\leftarrow cut off SS side

checked under microscope: diamond cut is good enough to show that the MgB_2 powders are infiltrated with Mg, forming a Mg/MgB_2 composite.

Took a polaroid, see next page!

▲ open small crucible

• cut 1: Mg to the rim, except small blowhole



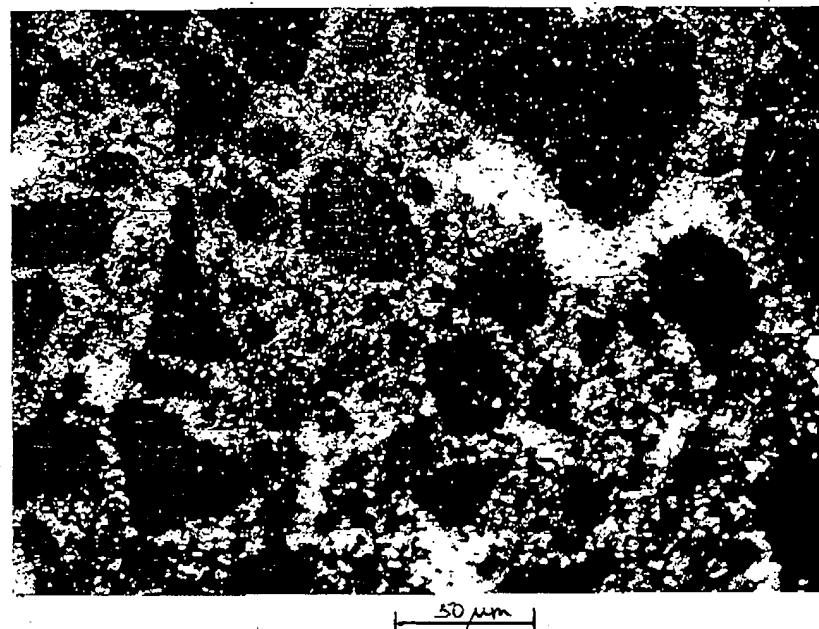
• cut #2 shows only powder, no infiltration

• possible problem: 1) leak at nut

2) bad seal by liquid Mg (unlikely)

3) premature solidification (high surface area)

discard sample.



Mg: white
MgB₂: black

$\frac{1}{3}$ Mg
 $\frac{2}{3}$ MgB₂

shorted here
continuous
current
path.

Polaroid: plug side of cut 2, very bottom of sample sample Mg/MgB₂ (L) 40x objective. (400x magnification)

- MgB₂ is agglomerated (size ~50-100 μm)
- Mg infiltrates as veins between agglomerates (veins: ~10-30 μm)
- many agglomerates are also filled with Mg (~1-5 μm)
- some show holes (pull out due to cutting?)

→ This is a fully infiltrated composite, with a few possible regions of porosity (may be due to closed porosity in original powders or infilling)

To Do: mount & polish this sample

machine steel off

measure conductivity vs T.

next experiments (possible directions)

Time: 0.100 ± 0.5%
Temp: 1500 ± 50°C
D: 15.25 mm ± 0.25 mm

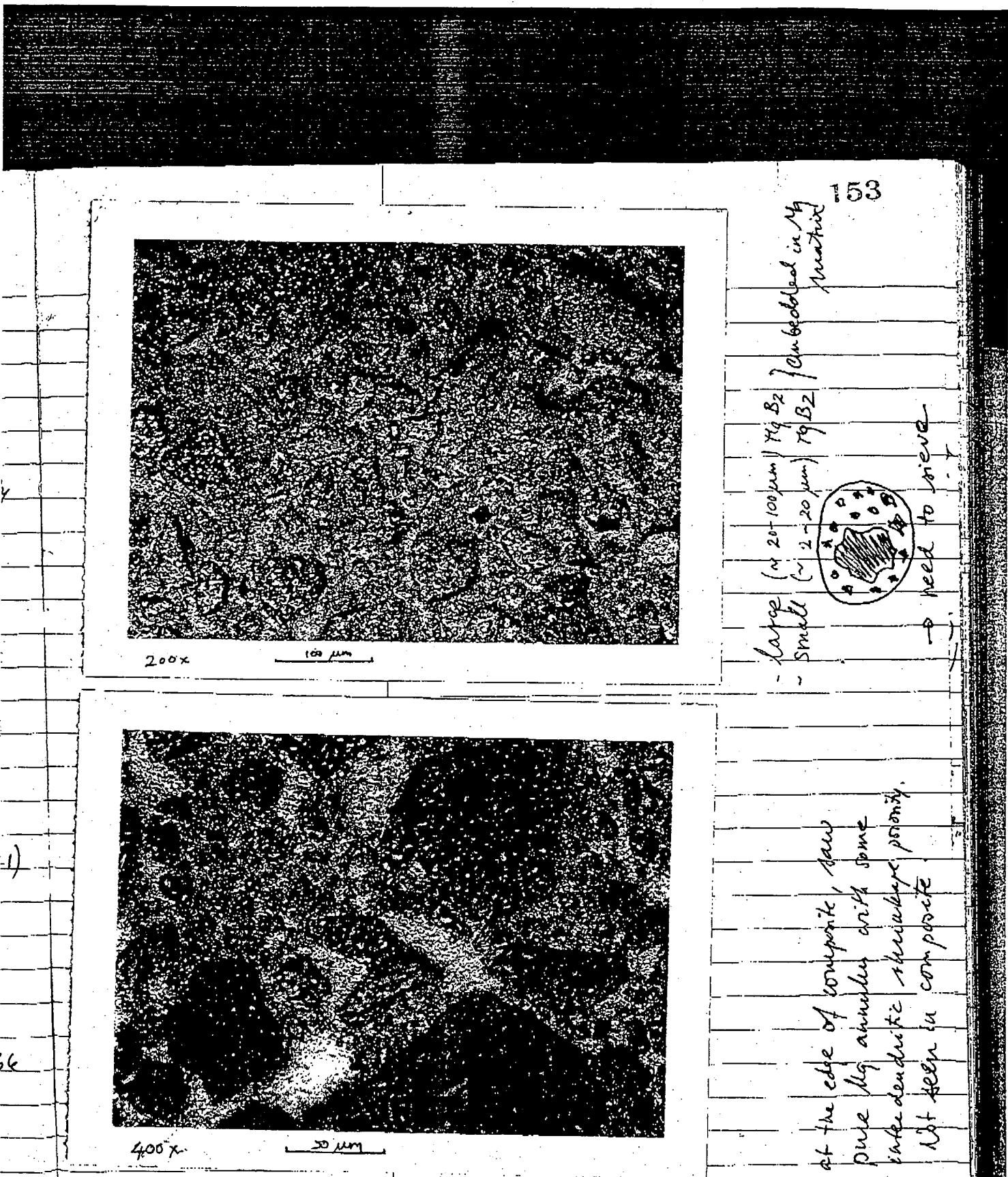
- does Hg wet MgB_2 ?
use steel crucible to encapsulate
 $MgB_2 + Hg$
also may show enhanced sintering due to Hg vapors
(check density of green compact with porosity)
- react B fibers in capsule, then infiltrate
- " B powders " " "
- infiltrate B fibers and then react in molten state
" B powders " " "
- infiltrate MgB_2 with Al } new crucibles
" Cu } or BN coated steel
- " " " "

- polishing some metallographic section (composite MMC)
 1. SiC paper with the
 2. 1 μm diamond in oil (0.05 Al_2O_3 grain up neutral)
 took 2 pictures (see next page)

- composite is successful: 1) full infiltration, no porosity
 2) no reaction between Hg & MgB_2
 3) no reaction of either phase w. crucible

Machine shop: asked for EDM of MMC!
 - welding of crucibles

Ordered: B powders, Tl_2O powders, more steel



Metals which can be infiltrated in centrifuge machine

	Tm	T ACS	
Cu	1088 (?)	100%	too hot!!
→ Ag	962	108%	
Al	662	65%	
Mg	650	39%	
Zn	420	28%	
Sn	232	16%	
(Brass 260 955K)		28%	
" 464 900		26%	
In	156	20%	
Pt	327	8%	
Ed	324	25%	
Ar	1064	73%	too late!!

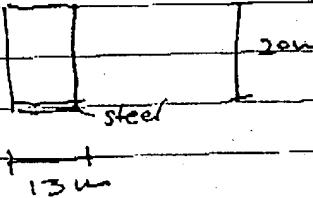
Mutual solubilities in Hg-X systems

in Hg in X intermetallic eutectic

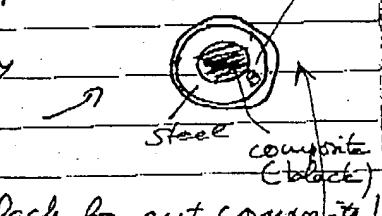
Hg - Hg	0	0	-	-
Hg - Nb	0	0	-	-
Hg - Co	0	0	HgCo ₂	635
Hg - Cr	0	0	-	no phase diag.
Hg - Fe	0	0	-	-
Hg - Be	0	0	1	
Hg - Nd	0	~2% (sat.)	4	588
Hg - Pd	0.23%	2.5at%	7	570
Hg - Pt	0	0	5	575
Hg - Ti	0.12%	1.5at%	2	651
Hg - Cu	0	6 at%	2	483
Hg - Au	0	huge	many	575

mass: 5.213 g

Received machined sample from machine shop. Machinist found composite to be much harder than stainless steel: bridle seems well bonded to matrix.

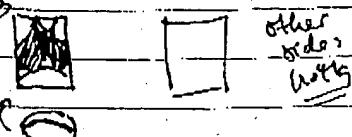


- Side of cylinders show a few pure Mg regions (shiny) ^{marked}
but mostly composite
- top part was faced \rightarrow
- bottom part is cut from diamond saw
 \rightarrow need to cut again with diamond saw
- The difference of color (gray for machined composite, black for cut composite) is probably due to smearing of Mg during lathe machining
- cut bottom slice (1.5 mm) with diamond saw (or 2 h!); other side looks nicely infiltrated, except for a large area of pure Mg. Unlike the other side, it is not black (could be due to corrosion etc.). Keep slice and mark it with a "B". Actually, due to methacrylate 1 mm in it produces black color
- it is probably etched by it!
- cut upper slice ()
- final dimensions: $h = 17.044 \text{ mm}$ $\rho = 12.778 \text{ g/cm}^3$ $r = 6.374 \text{ mm}$ $m = 4.378 \text{ g}$



$$f \cdot 2.63 + (1-f) \cdot 1.74 = 2.016 \quad f(2.63 - 1.74) = 2.611 - 1.74 = 0.874$$

30.5 vol% MgB₂



large regions of pure Mg, possibly displaced by falling clusters of Mg upon loading in crucible. In Mg region, volume factor is larger.



Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

**TRANSMITTAL
FORM**

(to be used for all correspondence after initial filing)

Total Number of Pages in This Submission	5	Attorney Docket Number	6513-DIV
--	---	------------------------	----------

ENCLOSURES (check all that apply)

<input type="checkbox"/> Fee Transmittal Form <input type="checkbox"/> Fee Attached <input type="checkbox"/> Amendment / Reply <input type="checkbox"/> After Final <input type="checkbox"/> Affidavits/declaration(s) <input type="checkbox"/> Extension of Time Request <input type="checkbox"/> Express Abandonment Request <input type="checkbox"/> Information Disclosure Statement <input type="checkbox"/> Certified Copy of Priority Document(s) <input type="checkbox"/> Reply to Missing Parts/ Incomplete Application <input type="checkbox"/> Reply to Missing Parts under 37 CFR1.52 or 1.53	<input type="checkbox"/> Drawing(s) <input type="checkbox"/> Licensing-related Papers <input type="checkbox"/> Petition <input type="checkbox"/> Petition to Convert to a Provisional Application <input type="checkbox"/> Power of Attorney, Revocation Change of Correspondence Address <input type="checkbox"/> Terminal Disclaimer <input type="checkbox"/> Request for Refund <input type="checkbox"/> CD, Number of CD(s) _____ <input type="checkbox"/> Landscape Table on CD	<input type="checkbox"/> After Allowance Communication to TC <input type="checkbox"/> Appeal Communication to Board of Appeals and Interferences <input type="checkbox"/> Appeal Communication to TC (Appeal Notice, Brief, Reply Brief) <input type="checkbox"/> Proprietary Information <input type="checkbox"/> Status Letter <input checked="" type="checkbox"/> Other Enclosure(s) (please identify below): Supplemental Response; Original, executed Declaration of David C. Dunand; Return receipt postcard
Remarks		
Authorization is hereby given to charge or credit Deposit Account No. <u>18-0882</u> for any fee deficiency or overpayment.		

Rodney D. DeKruif

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT

Firm	Reinhart Boerner Van Deuren s.c.		
Signature			
Printed Name	Rodney D. DeKruif		
Date	January 18, 2005	Reg. No.	35,853

CERTIFICATE OF TRANSMISSION/MAILING

I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as Express Mail, No. EV 531802408 US, in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the date shown below.

Signature			
Typed or printed name	Rodney D. DeKruif	Date	January 18, 2005

This collection of information is required by 37 CFR 1.5. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.



1 - 19 - 05

IFW

PATENT

I hereby certify that this paper or fee is being transmitted with sufficient postage as Express Mail, No. EV 531802408 US, in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on January 18, 2005.

Dated: January 18, 2005

BY: Rodney DeKruif
Rodney D. DeKruif

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: David C. Dunand)
)
Serial No: 10/680,639)
) Attorney Docket No. 6513-DIV
)
Filed: October 7, 2003)
)
For: SUPERCONDUCTING)
Mg-MgB₂ AND RELATED)
METAL COMPOSITES)
AND METHODS OF)
PREPARATION)

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

SUPPLEMENTAL RESPONSE

Dear Sir:

As a supplement to the Response filed on January 14, 2005, please enter and consider the original, executed Declaration of David C. Dunand, supporting patentability of the claimed invention. A copy of the same Declaration was previously filed.

Applicant believes no additional fees are required, but authorization is hereby granted to charge Deposit Account No. 18-0882 for any fee deficiency.

This application is now believed to be in condition for allowance. Action consistent therewith is respectfully requested. Thank you for your time and consideration.

Respectfully submitted,



Rodney D. DeKruif
Attorney for Applicant
Reg. No. 35,853

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- BLACK BORDERS**
- IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- FADED TEXT OR DRAWING**
- BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- SKEWED/SLANTED IMAGES**
- COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- GRAY SCALE DOCUMENTS**
- LINES OR MARKS ON ORIGINAL DOCUMENT**
- REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- OTHER:** _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.